

Ultrafoam Duplex Filter for Clean Coal Combustion

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Introduction

“Clean coal” technologies such as pressurized fluidized bed combustion (PFBC) and integrated gasification combined cycle (IGCC) require a hot gas filter to remove the corrosive and erosive coal ash entrained in the combustion gas stream. These hot gas filters, or candle filters, must be cost-effective while able to withstand the effects of corrosion, elevated temperature, thermal shock, and temperature transients. Ash loadings may range from 500 to 10,000 parts per million by weight, and may contain particles as fine as $0.2\ \mu\text{m}$ (8.0×10^{-6} in). The operating environment for the hot gas filter can range in pressure from 10 to 20 atmospheres; in temperature from 370 to 950°C (700-1750°F), and can be oxidizing or reducing. In addition, the process gases may contain volatile chloride, sulfur, and alkali species. Field testing of various commercially available porous ceramic filter matrices has demonstrated a loss of up to 50% of as-manufactured strength after 1000 to 2000 hours of exposure to these operating conditions, although full-scale elements have remained intact during normal process operation.

Ultramet has developed a new class of hot gas filter material, the Ultrafoam Duplex Filter (UDF), that offers lower backpressure, higher permeability, longer life, and high filtration efficiency in the PFBC and IGCC environments. Subscale UDF elements have undergone accelerated corrosion testing at temperatures up to 1300°C (2372°F) at Ultramet, and have been subjected to 2815 hours of exposure to hot PFBC gases in the Westinghouse Advanced Particle Filtration System at Brilliant, OH, without any loss in strength in either case. The UDF matrix demonstrated 100% particle capture efficiency of coal ash and had an initial pressure drop of 0.1 to 0.6 in·wc/fpm.

Oxidation resistance, corrosion resistance, thermal shock/thermal fatigue, C-ring strength, and creep strain of the CVD SiC foam filter body, and pressure drop and particle capture efficiency of subscale Ultrafoam Duplex Filter elements, are reported. Future work is targeted at the construction of full-scale hot gas filters, and will include further mechanical characterization and accelerated corrosion testing of subscale sections of the filter body, the membrane, and the composite duplex filter elements.

Objectives

The objectives of this advanced hot gas filter development effort are to:

- Design, develop, and demonstrate an advanced hot gas filter element (the Ultrafoam Duplex Filter) that is durable, corrosion-resistant, and resistant to damage from thermal fatigue, thermal transients, pulse cleaning, and mechanical ash bridging loads within a filter array subjected to a PFBC or IGCC environment.
- Assess the uniformity, repeatability, toughness, and damage tolerance of the UDF.
- Assess the steady-state filtration efficiency and pressure drop of the UDF.
- Assess the effects of the corrosion, thermal shock, and mechanical stresses typical of advanced coal-fired power systems on the strength and integrity of the UDF.
- Fabricate UDF elements and evaluate their performance in a simulated PFBC environment using the high-temperature, high-pressure (HTHP) fluidized-bed combustion (FBC) test vessel at the Westinghouse Science & Technology Center.
- Demonstrate that the UDF is a rugged, long-life, efficient advanced hot gas filter.

Technology

The Ultrafoam Duplex Filter is composed of a chemical vapor deposition (CVD) silicon carbide (SiC) reticulated open-cell foam filter body supporting a porous mullite membrane filter, as illustrated and shown in Figures 1A-B. The reticulated foam structure, shown in Figure 2, is 70-90% porous, enabling high permeability, while the three-dimensionally interconnected cellular lattice resists crack propagation. The CVD SiC material is fine-grained for maximum strength and corrosion resistance, and has no binders or impurities. The porous mullite membrane filter is impregnated into the outer surface of the CVD SiC foam filter element. The membrane bonds to the foam ligaments, while the foam ligaments reinforce and support the membrane. The mullite membrane, which acts as a barrier filter, is resistant to thermal shock and corrosion. The thin membrane minimizes pressure drop and thermal gradients. The composite UDF was designed to combine optimal material selection with functional structural design to maximize corrosion resistance, thermal shock resistance, strength, toughness, and life while minimizing pressure drop and weight.

The beneficial properties of the SiC foam filter body and the membrane barrier filter can be summarized as follows:

Ultrafoam Filter Body

- Structure: Reticulated open-cell foam with 3-D interconnected open porosity
- Material: Stoichiometric CVD SiC, with no impurities and very fine grains
- Tailored Properties:
 - 70-90% porosity
 - Pore size 10-100 pores per linear inch (ppi)
 - Controllable pore shape
 - 6.9-55.2 MPa (1000-8000 psi) uniaxial strength (a function of density)
 - Low pressure drop
 - Corrosion resistance
 - Thermal shock resistance
 - Damage tolerance

Membrane Barrier Filter

- Structure: Porous ceramic membrane
- Material: Mullite
- Tailored Properties:
 - 30-60% porosity
 - Controllable thickness
 - Controllable pore diameter
 - Integral part of Ultrafoam filter body; reinforced by foam ligaments
 - Low pressure drop
 - Corrosion resistance
 - Thermal shock resistance

Approach

The work to date has focused on demonstrating the feasibility of the UDF and then scaling up the UDF for continued performance assessment in several areas:

- Accelerated corrosion testing of the SiC foam filter body
 - Accelerated oxidation testing
 - Accelerated corrosion testing in NaSO_4
 - Thermal shock/thermal fatigue testing
 - Oxidation rate modeling
- Mechanical characterization of the SiC foam filter body
 - Room and elevated temperature C-ring strength
 - Elevated temperature creep strain
- Measurement of gas flow permeability and particle capture efficiency on UDF disk specimens
- Extended exposure of SiC foam filter body specimens to a simulated operating environment (Tidd exposure)

Results

Accelerated Corrosion Testing

Three types of accelerated corrosion testing were performed to characterize the durability of the SiC foam filter body. First, accelerated oxidation was evaluated by subjecting SiC foam specimens to 50, 100, and 200 hours of exposure in air at 800, 1000, and 1300°C (1472, 1832, and 2372°F). The thickness of the resultant surface oxide layers and the compressive strength of the foam specimens were measured after each exposure. Second, accelerated corrosion was evaluated by coating SiC foam specimens with a 10-wt% NaSO_4 solution (5,000 to 10,000 times the anticipated concentration in the projected use environment) and heat treating them in air at 800°C for 100 hours. Every 24 hours, the specimens were air quenched to room temperature and recoated with the NaSO_4 solution. The oxide layer thickness and foam compressive strength were measured after the test. Third, thermal shock/thermal fatigue response was evaluated by subjecting SiC foam specimens to a cycle consisting of rapid heatup to 1649°C (3000°F) in air via direct oxyacetylene torch

impingement, then air quenching to 600°C (1112°F). The cycle repeated every 6 seconds for up to 40,000 cycles, or a total of 240,000 seconds (4000 minutes, nearly 67 hours).

Figures 3A-B show the measured compressive strength of SiC foam specimens after various corrosive treatments, expressed as a function of relative density. The strength of the SiC foam filter body was shown not to be affected by accelerated oxidation, corrosion, or thermal shock/fatigue.

SiC typically forms a protective scale of SiO₂ upon coming in contact with most oxygen-containing atmospheres. The growth of the SiO₂ scale is known to follow a parabolic rate law. As described above, experimental data on the oxidation of SiC foam in air were obtained for temperatures of 800, 1000, and 1300°C and durations ranging from 50 to 200 hours, with the oxide layer thickness on the SiC foam specimens being measured as a function of time (exposure duration) and temperature. Additionally, a SiC foam specimen subjected to 2815 hours of Tidd plant exposure, described below, had shown approximately 1 μm (4 × 10⁻⁵ in) of oxide buildup, and this data point was also incorporated in the oxidation model being developed.

Using the relation

$$x = k_0 e^{-\frac{E_a}{RT}} t^{1/2} \quad (1)$$

where x is the oxidation rate, k_0 is the preexponential factor, E_a is the activation energy of the oxidation reaction, R is the universal gas constant, T is the temperature of exposure, and t is the time (duration) of exposure, and performing a least-squares fit of the experimental data (x , t , T) to the equation, the best-fit values for k_0 and E_a were determined to be 2.73 μm/hr^{1/2} and 10,600 cal/mol respectively. By combining these values with equation (1), the predicted oxide growth curves shown in Figure 4 were generated.

It should be noted that because the oxide scales were extremely thin (≤1.0 μm), the degree of uncertainty in the measurements (typically ±0.3 μm) is not insignificant. Additional long-term data are needed to confirm the model.

Mechanical Properties

The mechanical properties generated for SiC foam filter bodies included room and elevated temperature C-ring compressive and tensile strength and elevated temperature creep strain.

The C-ring strength tests were performed on SiC foam filter bodies without the membrane filter attached. Attachment of the membrane filter is anticipated to appreciably increase the strength of the UDF. The results of the C-ring tests, performed at room and elevated temperature (871 °C/1600°F), are shown in Table I. Additionally, the SiC foam filter body (without the membrane filter) has demonstrated C-ring strengths of >20.7 MPa (3000 psi) at process temperature. In open-cell foam structures, strength is typically a function of the material and the relative density (volume percent solid). Future work will be directed at determining the controlling factors of foam strength and characterizing the greater strength obtained from the addition of the membrane filter to the SiC foam filter body.

Elevated temperature creep strain in SiC foam specimens was measured for 400-450 hours at 843 °C (1550 °F) under four-point loads of 1.7 and 3.4 MPa (250 and 500 psi), per the matrix in Table II. Figure 5 shows representative results of this testing, indicating that the SiC foam did not experience elevated temperature creep.

Gas Flow Permeability and Particle Capture Efficiency

Gas flow permeability and particle capture efficiency were measured on UDF disk specimens incorporating various membrane filters. Figure 6 shows the results of the gas (air) flow permeability tests. All variations of the UDF disks had acceptably low pressure drops of 0.1 to 0.6 in-wc/fpm. Particle capture efficiency was measured by passing an air stream containing entrained ash from an advanced coal-fired power plant through a UDF disk specimen. For all specimens tested, 100% of the particles were captured. No particles were found downstream of the UDF disk.

Tidd Exposure

SiC foam filter body specimens were subjected to 2815 hours (117 days) of exposure to filtered PFBC process gases at temperature (843°C/1550°F) in the Westinghouse Advanced Particle Filtration System at the AEP Tidd demonstration plant in Brilliant, OH. The post-exposure residual strength of the SiC foam specimens was 24-32 MPa (3500-4650 psi), as measured by compressive C-ring testing at 843°C, which is equal to or greater than the as-fabricated strength. The SiC ligaments of the foam specimens were observed to have grown a 1- μm (4×10^{-5} in) layer of SiO_2 . Some fine ash was retained on the ligament surfaces. The oxidation behavior of the SiC ligaments over the life of a candle filter is unknown, although the model presented above predicts a SiO_2 layer thickness of $<3 \mu\text{m}$ (1.2×10^{-4} in) after 20,000 hours (833 days) of exposure at 843°C. The adherence or spalling of the oxide layer as a function of life and pulse cleaning frequency must be studied in future work.

Summary and Conclusions

- The SiC foam filter body has demonstrated substantial corrosion resistance, thermal fatigue endurance, and thermal shock resistance.
- The SiC foam filter body showed no change in strength after accelerated oxidation/corrosion trials.
- The SiC ligaments of the foam filter body form a coherent SiO_2 layer that increases following a parabolic rate law. After 2815 hours at 843°C in a PFBC environment, the SiO_2 layer was approximately 1 μm (4×10^{-5} in) thick.
- The SiC foam filter body has been fabricated to C-ring strengths of >20.7 MPa (3000 psi) at process temperature.
- The SiC foam filter body did not experience elevated temperature creep under four-point loading at 1.7 and 3.4 MPa (250 and 500 psi) for 450 hours at 843°C (1550°F).
- The residual C-ring compressive strength of the SiC foam filter body, after 2815 hours of field exposure, was equal to or greater than the as-fabricated strength.
- The Ultrafoam Duplex Filter showed acceptable gas flow permeability (pressure drop) and particle capture efficiency (barrier filtration) characteristics for advanced coal-fired hot gas filtration applications.
- The UDF has clearly demonstrated its potential as an advanced hot gas filter for use in advanced coal-fired applications.
- Further demonstration and characterization of UDF performance is required.

Future Work

- Fabrication will be scaled up to filter element sizes (lengths) of 1 to 1.5 meters.
- The effects of the corrosion, thermal shock, and mechanical stresses typical of advanced coal-fired power systems on the strength and integrity of the UDF will be assessed.
- The steady-state pressure drop (gas flow permeability) and filtration efficiency (particle capture) of the UDF will be assessed.
- UDF performance will be evaluated in a simulated PFBC environment using the high-temperature, high-pressure fluidized-bed combustion test vessel at the Westinghouse Science & Technology Center.
- The uniformity, repeatability, toughness, and damage tolerance of the UDF will be assessed.

Acknowledgments

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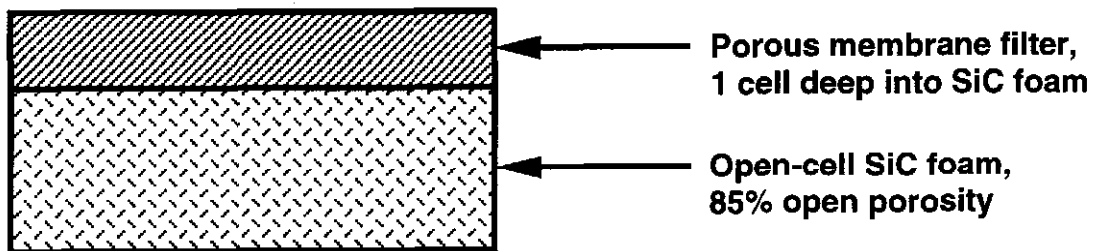


Figure 1A.
Schematic of Ultrafoam Duplex Filter structure

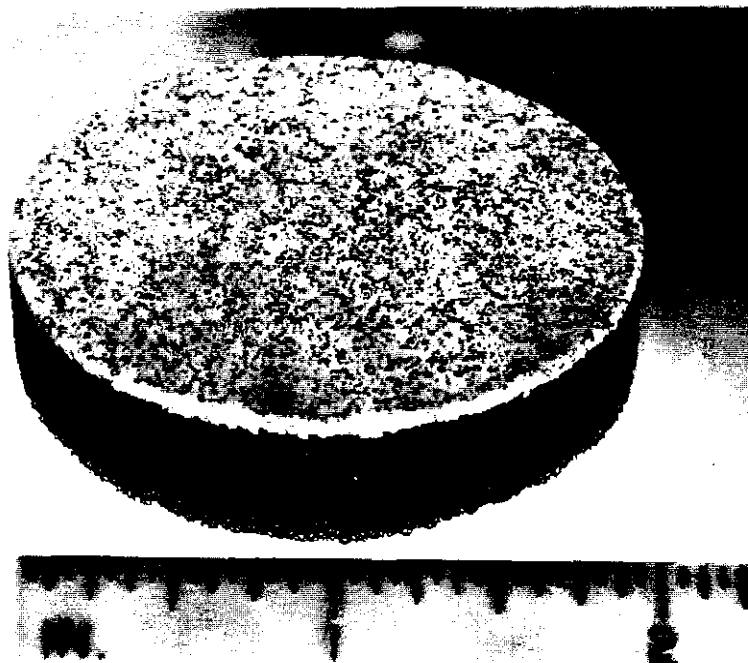


Figure 1B.
Ultrafoam Duplex Filter specimen

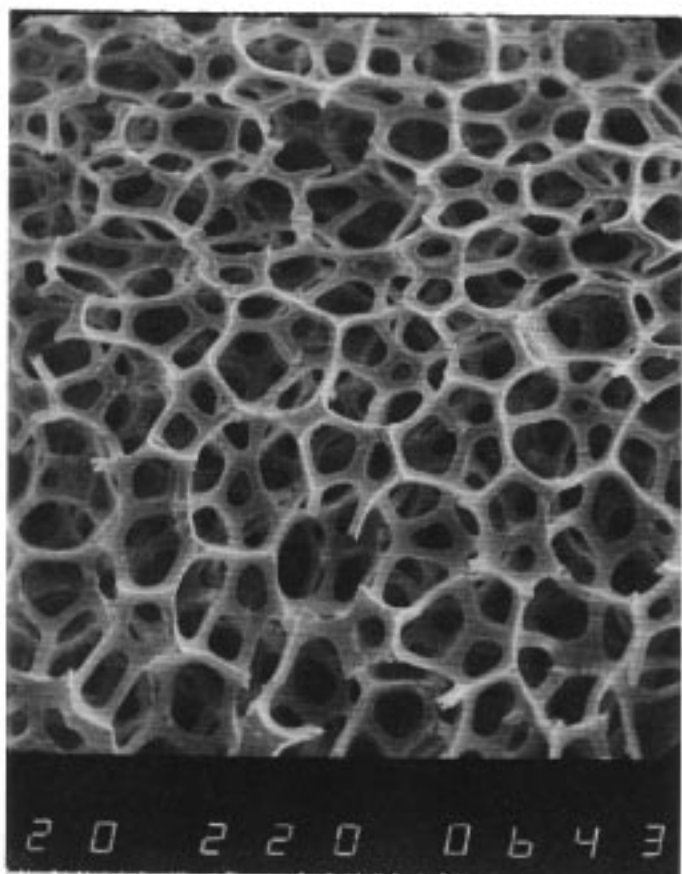


Figure 2.
SEM micrograph of open-cell foam microstructure (22×)

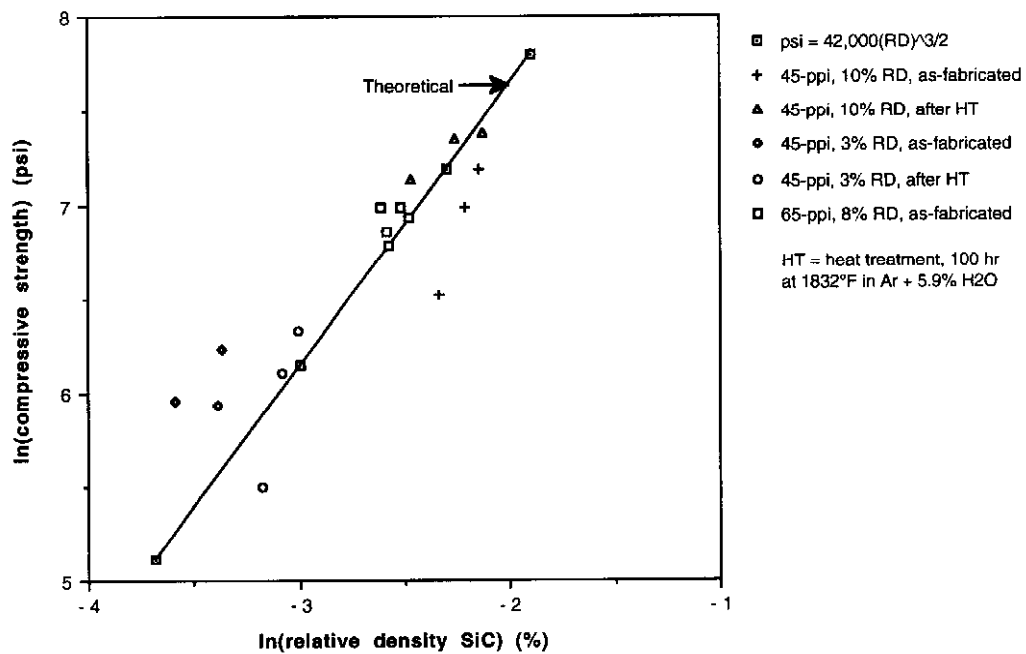


Figure 3A.
SiC foam compressive strength vs. relative density for as-fabricated and heat-treated specimens of various ppi and relative density

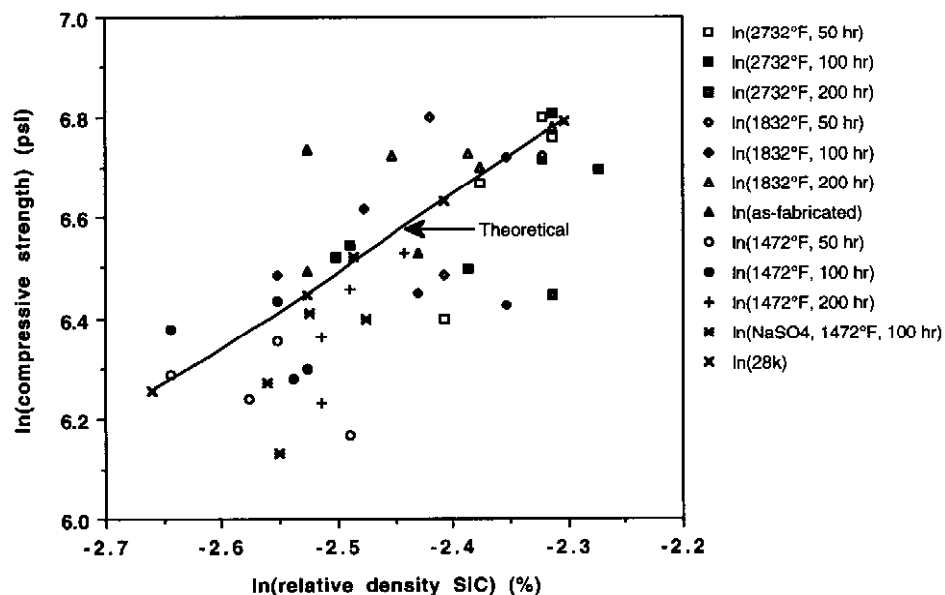


Figure 3B.
SiC foam compressive strength vs. relative density for 45-ppi specimens (as-fabricated, oxidation tested, and corrosion tested)

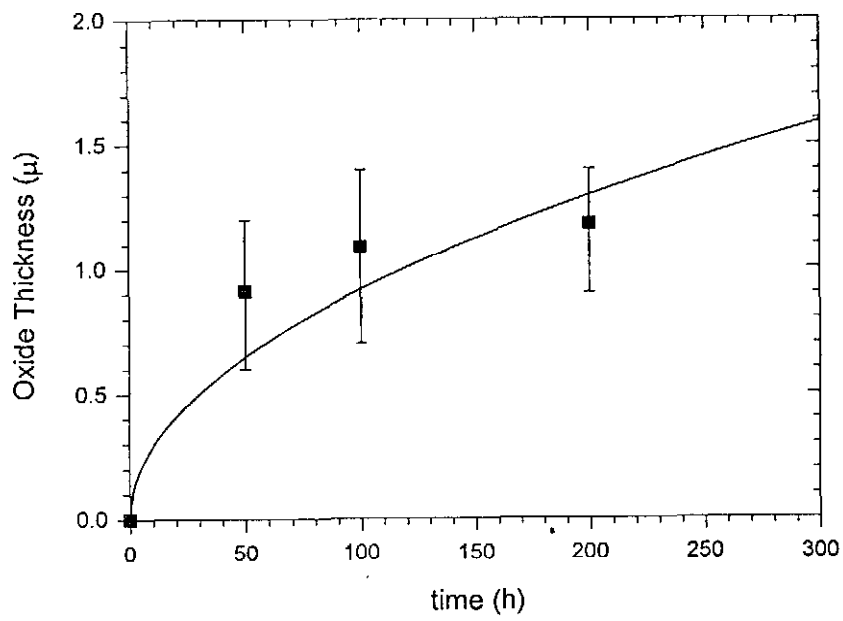


Figure 4.
SiC foam oxidation rate (oxide scale growth) in air at 1300°C. Shown: test data for 50, 100 and 200 hours of exposure, and model prediction.

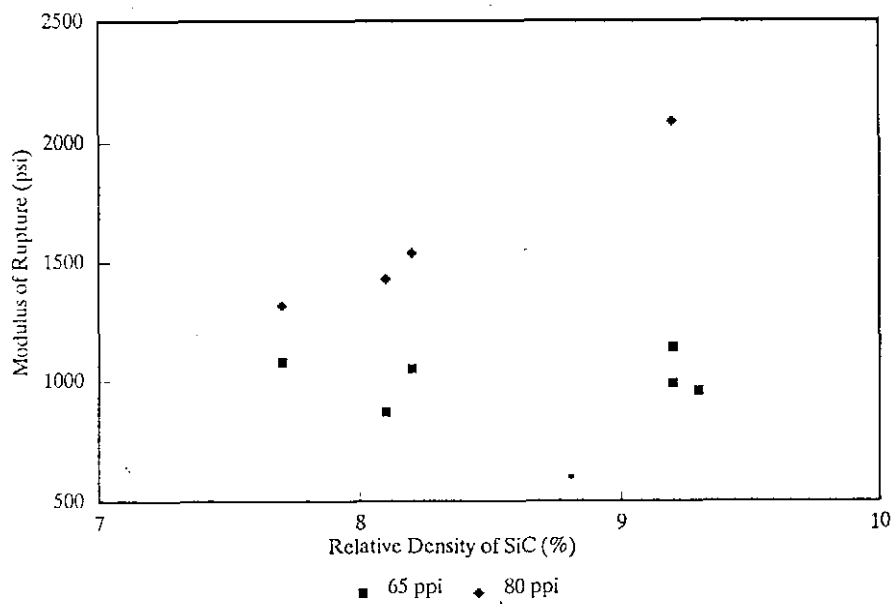


Figure 5.
SiC foam modulus of rupture vs. relative density from elevated temperature creep strain measurements

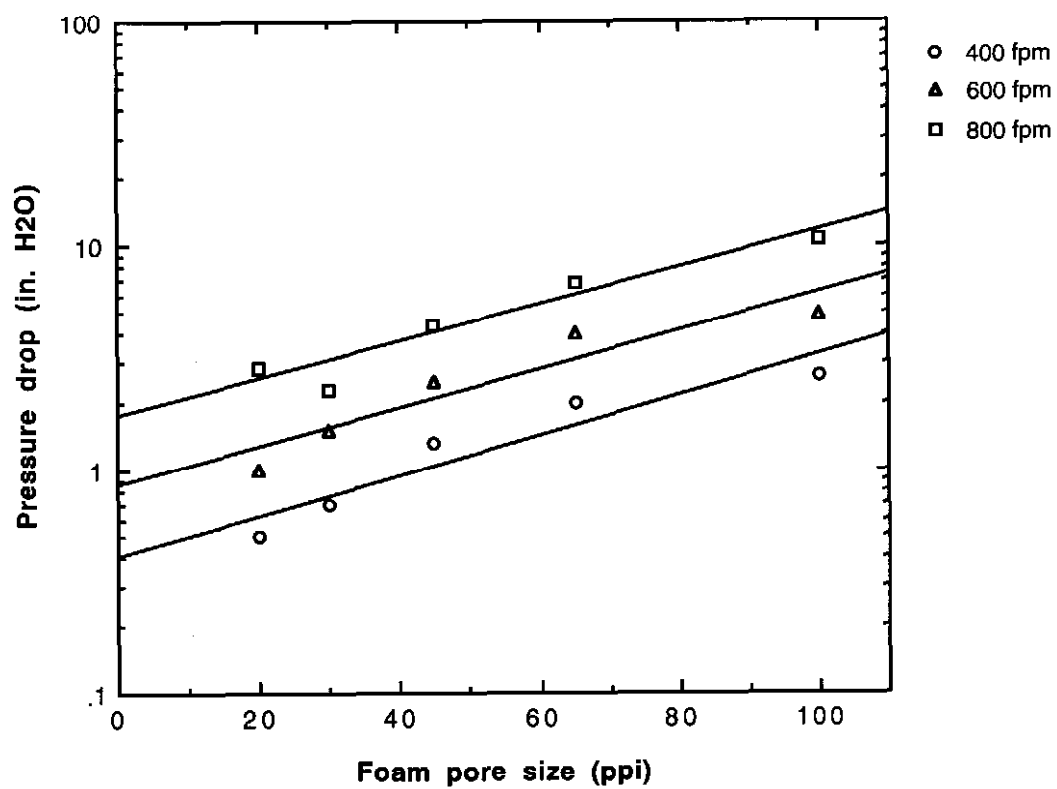


Figure 6.
SiC foam pressure drop vs. ppi at various air flow rates

Table I.
Bulk Strength of Various As-Fabricated SiC Foam Specimens (psi)

				C-Ring Compression		C-Ring Tension	
Sample	ID Number	Description	Wall Thickness, inch	RT	870°C	RT	870°C
A	6312-22-5 6312-21-3 6312-21-5 6312-21-2	45 ppi, 10% SiC	0.457-0.482	535±42	451±84	421±73	462±145
B	6312-32-3 6312-31-3 6312-31-4 6312-30-3	45 ppi, 14% SiC	0.462-0.496	416±116	407±104	527±97	319±8
C	6312-14-2 6312-16-4 6312-16-2 6312-17-1	65 ppi, 13% SiC	0.468-0.477	766±195	821±69	861±165	658±52
D	MIPS-1H30	100 ppi, 30% SiC	0.206-0.229	2433±382	3085±490	3053±702	3290±398

Table II.
Test Matrix for Elevated Temperature Creep Strain Measurement

Foam ppi	Relative density SiC (%)	Load (psi)
45	12.3	250
65	9.4	250
65	12.0	250
100	29.5	500